

Single Crystal Synchrotron X-Ray Charge Density Study of the Inverse Thermoelectric Clathrate $\text{I}_8\text{As}_8\text{Ge}_{38}$

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Beamline(s): **X3A1**

Inorganic clathrates have recently been intensively investigated as potential thermoelectric materials. The clathrates appear to fulfil many of the material requirements for a good thermoelectric exhibiting low thermal conductivities achieved by the efficient phonon scattering by anharmonically vibrating guest atoms and an electronic conductivity that is not affected by the atomic "rattling". Most of the clathrates studied so far have substitution of framework group IV atoms with atoms to the left in the periodic table. To compensate for the lack of framework electrons in the substituted structures, cationic guest atoms are incorporated into the cages of structures. In order to better understand the many effects, which contribute to the thermoelectric properties, we have synthesized a number of inverse type I clathrates. These structures have framework substitution with atoms to the right in the periodic table, and anionic guest atoms in the cages. Our initial efforts have focused on $\text{I}_8\text{As}_8\text{Ge}_{38}$ where extensive single crystal diffraction data have been collected at 20, 100 and 300 K at X3A1 using a CCD detector (20 K: $a = 10.603(1) \text{ \AA}$, $V = 1192.1(1) \text{ \AA}^3$, $V_{\text{crystal}} = 0.00012 \text{ mm}^3$, $N_{\text{meas}} = 39122$, $N_{\text{unique}} = 963$, $R(\text{int}) = 0.037$, $R_{F2} = 0.0305$ (spherical atom model)). Since the structure is disordered on both the framework and the guest atom positions it has not been possible to model the electron density with multipole models. Instead we have used the maximum entropy method with a non-uniform prior distribution calculated from the spherical atom model. The corresponding maximum entropy deformation density provides a sensitive tool for studying the disorder and the charge redistribution upon formation of the solid.

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